communications earth & environment

ARTICLE

https://doi.org/10.1038/s43247-023-00723-7 OPEN

Heavy metal concentrations in rice that meet safety standards can still pose a risk to human health

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Long-term consumption of rice containing heavy metal(loid)s poses significant risks to public health, which can be scientifically evaluated through food safety assessment. However, spatial variability and uncertainty in exposure parameters are generally neglected in existing food safety assessment standards. This study focused on rice consumption in 32 provinces of China, and extracted 3376 data points of five heavy metal(loid)s (cadmium, arsenic, mercury, lead, and chromium) and two nutrient elements (copper and zinc) from 408 articles. Probability and fuzzy methods were integrated to cope with the spatial variability or uncertainty and more accurately evaluate the risk. The results demonstrated that long-term consumption of rice that meets the national food safety standards still can cause non-negligible health risks, particularly for children and toddlers with chronical exposure. Arsenic and Cd were found to be the most critical elements, which contribute to 64.57% and 22.38% of the overall human health risk, respectively. Fuzzy assessment indicated that the score in northern China is approximately eight folds of that in southern China, indicating that northern rice has lower risks and better nutrition. Our results demonstrate that the food safety standards need to be tailored according to local conditions with more specific receptor parameters and risk acceptance.

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Food safety is of great concern for human health and social well-being[1]. Although dietary diversity can reduce health risks to some extent[2], staple foods remain the key source of nutritional intake. Rice is the staple food for over half of the world's population[3]; however, it is also more vulnerable to pollution than other crops. For instance, the heavy metal(loid) accumulation capacity of rice is approximately three times that of wheat[4]. The toxicity, bioaccumulation, and potential toxic effects of heavy metal(loid)s may pose significant risks to human health[5]. To minimize these health risks, international organizations and national administrations have set the maximum acceptable concentration (MAC) of heavy metal(loid)s for rice. For example, the MACs of arsenic (As) and copper (Cu) set by the United Nations Food and Agriculture Organization and World Health Organization are 0.2 and 10 mg kg^{-1}[6], respectively. However, heavy metal(loid)s at concentrations below the MAC can still present health risks. Some studies have demonstrated that long-term exposure to low concentrations of As can cause non-carcinogenic diseases such as hypertension, neurological disorders, and even cancer[7]. In addition, exposure parameters vary with age, body weight, and region, leading to susceptible populations with higher risks[8,9]. Therefore, health risk assessments need to consider various factors such as body weight, age, dietary habits, and long-term intake besides MAC.

Health risk assessments are vital for a full understanding of public health status. Researchers have developed various evaluation methods such as in vitro digestive system[10], animal[11], and intestinal cell[12] models. However, these methods can hardly be adopted widely due to the impacts of human disturbance and ethical issues. As an alternative, human health risk assessment (HHRA) has become one of the most widely used methods, and has been adopted by many countries and international organizations[13]. This method can be used to flexibly select location-specific parameters with unified international reference standards. It can generate the most comparable evaluation results among all the currently used methods since it has been used in numerous studies. Although several studies have evaluated heavy metal(loid) pollution in rice, they were conducted mainly in local regions and cities[14–19]. Only a few nationwide surveys have been reported (China[20], Brazil[21], Spain[22], Kuwaiti[23], the United States[24], and several Southeast Asian countries[25]) approximately a decade ago. However, these studies simplified the calculation by using unified body weight and rice intake. As a result, the status of receptors exposed to heavy metal(loid)s through rice consumption, and the impact of receptor and regional differences on health risks remain unclear. In addition to heavy metal concentrations, HHRA also considers dietary habits and receptor differences, which can facilitate more accurate evaluation of health risks.

However, the HHRA evaluation results can be greatly affected by parameter uncertainty. Ignoring the uncertainty in health risk assessments may overestimate or underestimate the health risks, which may lead to improper decision-making[26]. Uncertainty can be classified as aleatory and epistemic uncertainty. In HHRA, aleatory uncertainty is caused by random changes in pollutant concentration, body weight, and daily intake. Epistemic uncertainty is due to the lack of data and ambiguity in risk perception among different assessors. Monte Carlo simulation has been demonstrated as one of the most useful methods to solve the problem of aleatory uncertainty with the availability of sufficient data to estimate the probability distribution of parameters[27]. Additionally, fuzzy analysis is a powerful tool to manage the fuzzy linguistic variables of an assessor and other epistemic uncertainties via fuzzy sets and membership functions[28]. Therefore, the integration of probability and fuzzy methods can effectively reduce the impact of uncertainty and quantify the potential health risks caused by long-term exposure to heavy metal(loid)s. Research in some other fields, such as contaminated site remediation, river pollution risk analysis, and water resource management[29–31], has provided good reference for food safety studies.

Although many studies have examined the heavy metal(loid) pollution in food, public concern about food safety has risen to an unprecedented level. Existing not fully elucidated the risk posed by rice intake due to the use of uniform parameters (such as body weight of 70 kg). In this study, we analyzed the heavy metal(loid) concentrations in commercial rice from various provinces of China and identified the spatial distribution of risk. The probability of health risks in the populations of 32 provinces was quantified by refining the parameters, which could illustrate the impact of receptor differences and dietary habits on the risk. The study aims to (1) accurately identify the critical receptors in different provinces and corresponding probability of the health risk to exceed the threshold, (2) evaluate the contribution rates of five heavy metal(loid)s (Cd, As, Hg, Pb, and Cr) in different provinces to health risks, and (3) clarify the mismatch between current national food safety (NFS) standards and the HHRA system. The results revealed the mismatch between NFS standards and actual human health risks, and indicated that the evaluation of heavy metal(loid) pollution risk in rice should be combined with studies of heavy metal(loid) concentrations and characteristics of the exposed population to obtain more accurate results, which may provide important implications for the formulation or tailoring of food safety standards.

Results

Heavy metal(loid) pollution in commercial rice. The average Cd, As, Hg, Pb, and chromium (Cr) concentrations in rice were 0.068, 0.021, 0.007, 0.065, and 0.121 mg kg^{-1}, respectively, which were far lower than the NFS standards (Cd = 0.2 mg kg^{-1}, As = 0.2 mg kg^{-1}, Hg = 0.02 mg kg^{-1}, Pb = 0.2 mg kg^{-1}, and Cr = 1.0 mg kg^{-1}, respectively). The maximum Pb, As, Hg, and Cr concentrations were 1.455, 1.1, 2.8, and 1.05 times of their risk thresholds, respectively. The average Cu and Zn concentrations in commercial rice were 2.31 and 15.429 mg kg^{-1}, respectively, indicating a good nutritional status.

The average value of the SFPI index followed the order of Pb (0.639) > Hg (0.595) > Zn (0.441) > Cu (0.437) > Cd (0.328) > As (0.205) > Cr (0.181). The results showed that the risk of heavy metal(loid) contamination in commercial rice was lower than the risk threshold (1.0). The SFPI values of Cd, As, Hg, Pb, Cr, Cu, and Zn in commercial rice ranged from 0.037 to 0.655, 0.058 to 1.1, 0.073 to 2.802, 0.441 to 1.5, 0.008 to 1.05, 0.187 to 1.35, and 0.199 to 0.882, respectively (Supplementary Fig. 5). The Hg concentrations in rice from Shaanxi, Guizhou, Jilin, Guangdong, and Hunan were 2.80, 2.20, 1.26, 1.07, and 1.01 times of those in the NFS standards, respectively, indicating that the food safety level of rice was not adequate. In addition, Pb concentrations in rice from Jiangsu, Anhui, Tianjin, Jilin, and Liaoning were 1.26–1.5 folds of those in the NFS standards. The As concentration in Taiwan and the Cr concentration in Sichuan were higher than their respective MACs. These results indicated that commercial rice in these provinces was contaminated by various heavy metal(loid)s. The relatively high Cu concentration in Guizhou rice indicated that the nutritional value of rice may be affected.

Risk status evaluated based on probability analysis. The average HQ of critical receptors followed the order of As (0.67) > Cd (0.65) > Cr (0.38) > Pb (0.16) > Hg (0.15) (Supplementary Table 5). For most provinces, the HQs of As and Cd were much higher than those of other three heavy metal(loid)s. The HQs of As in the three northeastern provinces were 10.20, 4.04, and 3.25 folds those of Hg, Pb, and Cr, respectively. The HQ of Cd was 2.50, 2.29, and 2.15 folds that of Hg, Pb, and Cr in the main rice-producing and rice-consuming areas (Hunan, Hubei, Jiangxi, and Guangxi) of southern China, respectively. In central and southern China, the NCR indicators (HQs) for As and Cd exceeded 1.0. The average CR for critical receptors was higher for As than for Pb. Overall, the mean CR values were all below 1×10^{-4}, which were within the acceptable risk range. In addition, regardless of the type of risk, the risk values were higher for children and toddlers than for adults.

When exposed to a toxic substance, the critical receptors with the most obvious response were the representative group in the exposure assessment (Supplementary Fig. 6). In general, young people (<18 years) were the critical receptors in all provinces. However, the specific age groups of the critical receptors were different in different provinces. The critical receptors were children (5–12 years) in about two-thirds of the provinces and toddlers (2–5 years) in the remaining provinces. The critical receptors in the central rice-producing provinces tended to be toddlers.

We plotted the cumulative distribution function of heavy metal(loid) exposure in rice (Supplementary Fig. 7). There was substantial diversity in the health risks for specific critical receptors in various provinces. In Gansu, Guizhou, Ningxia, Sichuan, Chongqing, and Taiwan, there were much higher health risks than in other provinces. The main risk in these six provinces was generated by As. The risk caused by Cd could not be ignored because its probability of exceeding the risk threshold for 20 provinces ranged from 0.003 to 0.992. Hg and Pb contamination affected three and six provinces, respectively. In contrast, the risk in Henan province was zero and there was no health effect on critical receptors due to heavy metal(loid)s. The cumulative effect of heavy metal(loid) exposure also showed significant differences among different provinces. The growth rate and distribution of the cumulative distribution function of Cd, As, Hg, Pb, and Cr varied significantly among different provinces. For example, the critical receptors in Hubei and Hainan were toddlers, and there was no significant difference in their BW and IR. However, the risk accumulation rate of Cd in Hubei was greater than that in Hainan due to the 2.5-fold difference in rice Cd concentration (yellow solid lines of HB1 and HN2 in Supplementary Fig. 7). In addition, the As concentrations in Guangdong and Jilin rice were similar (0.031 mg kg^{-1}), and the critical receptors were the same. Because the Guangdong population had 1.75 times rice intake of the Jilin population, Guangdong had a faster risk accumulation rate of As than Jilin (purple dotted-dashed lines of GD and JL in Supplementary Fig. 7).

The P_{NCR} values across provinces were calculated based on the HQ of the critical receptors in each province. The excess probability of all NCR indicators is shown in Fig. 1. The P_{NCR} varied significantly among different provinces (ranging from 0.005 to 0.997). The risk was significantly higher in central China than in other regions. The NCR increased gradually from the west to the east and from the north to the south. There were also provincial differences in the probability of NCR exceeding the risk threshold. The P_{NCR} values in 23 provinces were related to As. The P_{NCR} values in Guangdong, Guangxi, Hunan, and Jiangxi were related to Cd. Cr dominated the P_{NCR} in Yunnan and Guizhou. Although the average heavy metal(loid) concentrations in most provinces were lower than their respective MACs, the health risks caused by long-term exposure to relatively high levels of heavy metal(loid)s, particularly for sensitive groups (such as toddlers and children), may still be significant.

Arsenic and Pb were the CR assessment targets of ingested rice. The distribution of P_{ILCR} in each province is shown in Fig. 2. In 24 provinces, there was no unacceptable CR, and the P_{ILCR} was zero. In contrast, the CR in central and western China was slightly higher, with a mean P_{ILCR} of 0.413. The unacceptable CR in Taiwan was due to the excessive As concentration in rice. The As concentration was critical for determining the CR in all provinces due to its high carcinogenic toxicity (Supplementary Fig. 8). Long-term exposure to As, even at levels lower than the MAC, presented a significant CR. In contrast, Pb was not linked to any significant CR in the 32 provinces.

The total health risk was determined from the combined effect of various heavy metal(loid)s. Figure 3 shows the prominent contribution of As in northern China, with contribution rates ranging from 52.55 to 100%. There were obvious differences among provinces in southern China. The Cd concentration had the greatest contribution in Hunan, Jiangxi, Guangdong, and Guangxi, accounting for 51.60%, 97.48%, 44.31%, and 49.88% of the overall health risk, respectively. The contribution of Cr was the greatest in Guizhou and Yunnan (39.59% and 85.06%, respectively). Arsenic was the most significant contributor to human health risks, with an average contribution of 64.57%, followed by Cd with an average contribution of 22.38%. The Hg concentration only had a minuscule contribution to human health risks, with a contribution of merely 1.53%.

To ensure nutritional safety, the high P_{NV} between Cu and Zn was selected as the critical criterion for determining the impact on the nutritional value of rice. The results showed that the nutritional value of rice was not affected in most provinces, and the corresponding P_{NV} was zero. The final P_{NV} values of Anhui, Guangdong, and Inner Mongolia were 0.043, 0.095, and 0.033, respectively (Supplementary Figs. 9 and 10), confirming that the concentrations of nutrient elements in rice in these provinces were at a risk of exceeding the nutrient limits.

Rice quality score evaluated based on fuzzy analysis. Social demand for food is based on not only safety but also nutrition. Therefore, a comprehensive method is needed to assess both the heavy metal(loid) pollution level and nutritional value of rice. Here, we used a fuzzy analysis to integrate the P_{HR} and P_{NV} obtained from a probability analysis and finally obtained a comprehensive and specific RQHM score.

The P_{HR} and P_{NV} values of Heilongjiang Province were 0.265 and 0, respectively. The P_{HR} was mapped to the fuzzy membership function as shown in Supplementary Fig. 2. The critical level of health risk could then be described as partially L (${\mu^L}_{HR} = 0.175$) and partially LM (${\mu^{LM}}_{HR} = 0.825$), and the critical level of nutritional value was L (${\mu^L}_{NV} = 1$). Therefore, two different combinations of the health risk and nutritional value would affect the critical level. The fuzzy AND operator connects health risk and nutritional value effects. The fuzzy rice safety quality level can be determined according to the generated fuzzy rules as shown in Supplementary Table 4. For example, when the health risk was "L" (${\mu^L}_{HR} = 0.175$), and the nutritional value impact was "L" (${\mu^L}_{NV} = 1$), the RQHM was identified as "excellent (E)" (${\mu^E}_{RQHM} = 0.175$) (Supplementary Fig. 3). The different RQHM levels were aggregated into a shape representing the final fuzzy RQHM using the fuzzy OR operator. The RQHM in Heilongjiang Province was determined by calculating the centroid of the final shape (Fig. 4). Heilongjiang Province scored 72.72. Although the nutritional value of rice in Heilongjiang Province was good, and the heavy metal(loid) concentrations in rice also met the NFS standards, the health risks posed by Cd and As could not be accepted. The same fuzzy method was applied to various provinces in China to obtain the quality score of rice safety (Supplementary Fig. 11).

A high score indicates a high safety level for the rice and a lower risk to human health. The scores indicated good safety and

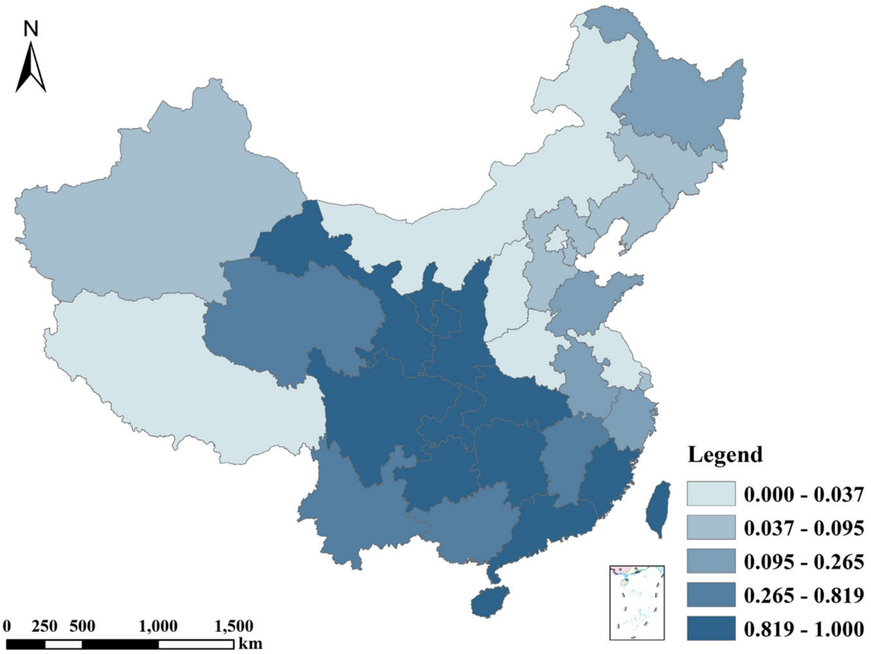

Fig. 1 Probability distribution of the non-carcinogenic health risk (NCR) exceeding the risk threshold (P_{NCR}). [Note: Regardless of the type of heavy metal(loid), the highest P_{NCR} in each province was taken to indicate the highest possible NCR. The shade of blue indicated the probability of non-carcinogenic risk exceeding the standard. The darker the blue, the higher the probability].

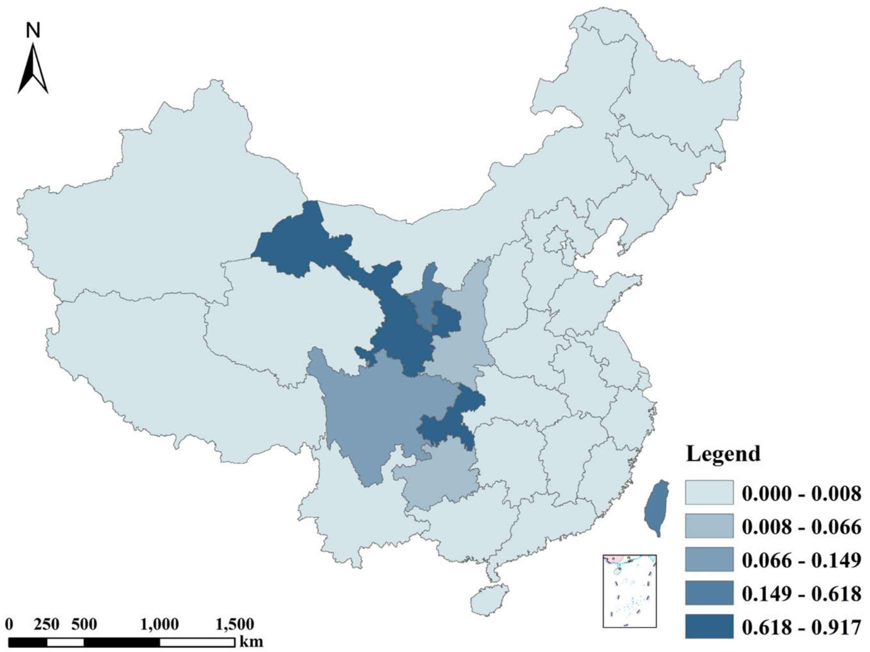

Fig. 2 Probability distribution of the carcinogenic health risk (CR) exceeding the risk threshold (P_{CR}). [Note: regardless of the type of heavy metal(loid), the highest P_{CR} in each province was taken to indicate the highest possible CR. The shade of blue indicated the probability of carcinogenic risk exceeding the standard. The darker the blue, the higher the probability].

quality of rice in northwest and northeast China (Fig. 5). Therefore, no measures are necessary to control heavy metal(loid)s in these areas. The scores in the central and western regions of China were not high, ranging from 46.60 to 81.07 (Supplementary Table 6). High As concentrations can cause significant health risks to sensitive populations. These provinces should make efforts to further reduce the heavy metal(loid) concentrations in rice and encourage producers and consumers to integrate heavy metal(loid) removal technologies into rice production and cooking process. The scores in southern China indicated that rice quality needs to be improved, with the lowest score being 10.83. The high Cd and Cr concentrations pose a significant NCR to sensitive people. Hainan, Guangxi, and Hunan are the main rice-producing areas of China. In addition to the approaches mentioned above, risk control measures in these provinces can be started from management of the pollution source, such as farmland rehabilitation and planting rice varieties with low accumulation of heavy metal(loid)s. In other major rice

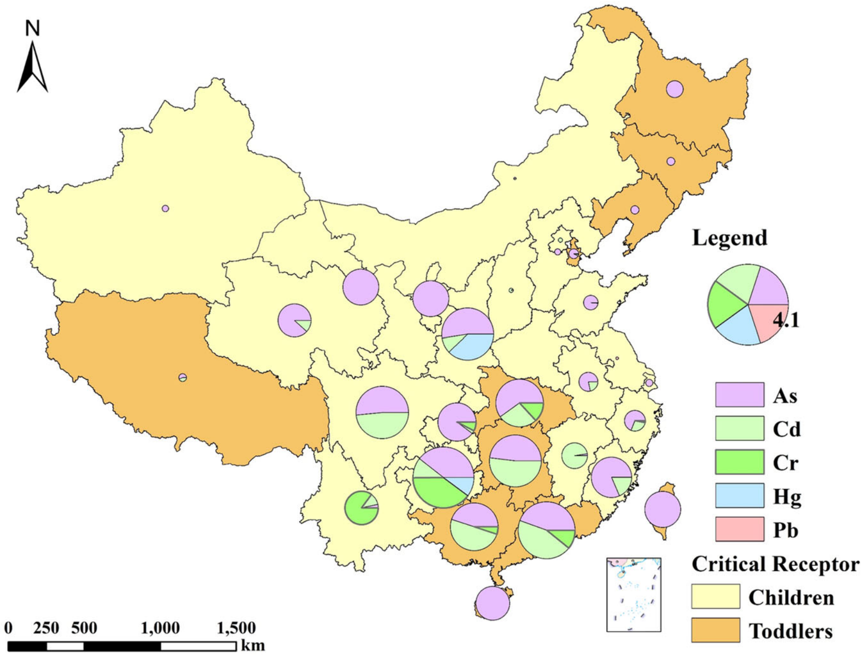

Fig. 3 Probability distribution of ultimate health risks and the contributions of Cd, As, Hg, Pb, and Cr to human health risks. [Note: The pie chart showed the contribution of five heavy metals(loid) to the health risk. The large (small) sector area indicates the high (low) risk contribution. A complete pie chart indicated a contribution of 100%. The size of the pie chart represented the sum of the probability of five heavy metal(loid)s exceeding the standard. The following abbreviations are used: Arsenic (As; purple), Cadmium (Cd; aqua), Chromium (Cr; green), Mercury (Hg; sky blue), Lead (Pb, pink)].

consumption areas such as Guangdong, residents should adjust their dietary structure to reduce their rice intake. At the same time, rice could be imported from places with lower heavy metal(loid) concentrations, such as northeast China. In provinces with low scores, there was a need to reduce the heavy metal(loid) concentrations in rice, thereby reducing human health risks.

Discussion

Differences in health risks among receptors. Our results are consistent with the overall trend of most other regional studies, including those of the elements posing the greatest threat to Chinese residents[32–35]. In some areas, severe Cd pollution in farmland poses a health risk[36]. In our study, Cr, Hg, and Pb did not pose significant health risks in most cases. Previous studies have concluded that these heavy metals pose health risks in specific regions[37]. These different results may be due to different research objects and exposure parameters. This study focused on commercial rice rather than locally grown rice, and derived appropriate exposure parameters for various age groups in 32 provinces.

Children and toddlers can be exposed to serious health risks. The HQs of young people in this study were ~1.1–1.5 folds those of adults due to differences in body weight and intake between the age groups, which is in good agreement with the predictions in other studies[32,35,38]. The average daily intake per unit body weight of the four intake groups follows the order of children (9.15 g kg^{-1}) > toddlers (9.00 g kg^{-1}) > teenagers (7.14 g kg^{-1}) > adults (4.45 g kg^{-1}) (Supplementary Table 1). Theoretically, changing the dietary structure can reduce health risks[39]. Taking wheat, potato, or corn with low heavy metal(loid) contents instead of rice as the staple food can reduce the total intake of heavy metal(loid)s, thereby reducing health risks; however, it remains a challenge to change the diet to reduce risks. For Chinese consumers, rice will remain the staple food for a long time, and people are still lack of awareness of the link between food consumption and health. Obviously, the physical inadequacy of children and toddlers should be taken into account, who are more likely to suffer from the toxic effects than adults when exposed to heavy metal(loid)s due to their high exposure frequency, smaller body size, and poor tolerance of heavy metal(loid)s[40]. Through multiple exposure routes, heavy metal(-loid)s have greater cumulative effects on young people than on adults. Therefore, the government should focus on controlling As and Cd levels, as well as pay attention to Cr, Pb, and Hg levels, to reduce the risk to children and toddlers from food intake and more comprehensively protect public diet health. Our results suggest that Hunan, Sichuan, and Guizhou Province should reduce mining intensity, monitor irrigation water quality in paddy fields, and plant rice varieties with low accumulation of heavy metal(loid)s, while Guangdong and Chongqing Province should import rice with lower heavy metal(loid) concentrations to protect human health.

Spatial transfer of health risks. The risks arising from the consumption of commercial and locally grown rice are not always the same. Due to the impact of human activities, there has been a spatial transfer of risks[41]. Because some risks of heavy metal(loid) exposure originate from rice imported from other provinces, the risk contribution of heavy metal(loid)s is not completely consistent with the local heavy metal(loid) pollution status. According to previous studies of rice, the Cd concentration in south China, the As concentration in northeast China, and the Cr concentration in Sichuan exceed the NFS standards[42]. However, our results showed that the intake risks in the main rice-producing areas, such as Hubei, Jiangxi, Guangxi, and Heilongjiang, were within the acceptable range. In general, rice production and consumption differ among provinces, and the inter-provincial supply and demand relationship of rice determines the level of inter-provincial trade of rice and risk transfer. Differences in IR and BW among different populations also lead to different risk profiles even with the consumption of rice at the same level of contamination. Rice without heavy metal(loid)

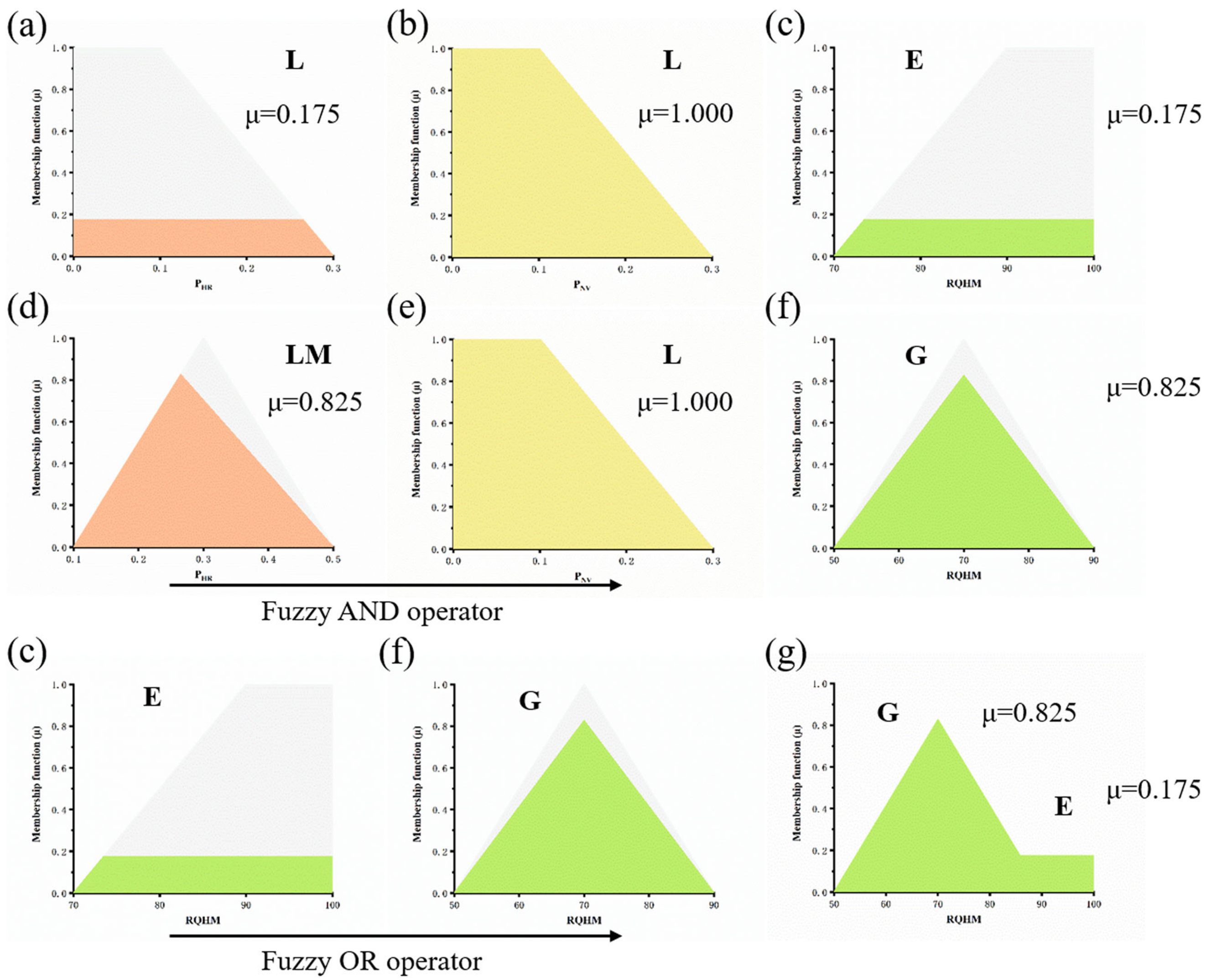

Fig. 4 Fuzzy inference process in Heilongjiang Province. [Note: **a**, **d**: health risks identified using health risk guidelines (pink); **b**, **e**: nutritional value impact (yellow); **c**, **f**, **g**: the rice quality–heavy metal(loid) (RQHM) score (green). The fuzzy AND operator of **a** and **b** and of **d** and **e** were used to obtain **c** and **f**; the fuzzy OR operator of **c** and **f** was used to obtain (**g**); μ indicated critical level, which was obtained by mapping the exceeding probability to the fuzzy membership function. The pale gray shading represented the initial fuzzy membership function, and the bright shading represented the fuzzy graph obtained by mapping the critical level into the fuzzy membership function].

intake risk in the area it is originally grown could also cause intake risks due to changes in the intake population. The boom in domestic and international trade has accelerated the transfer of such risks. E-commerce has proliferated in the past decade, and online shopping has become more convenient[43]. Furthermore, the Covid-19 epidemic has encouraged online shopping. Frequent shopping or trade behaviors induce inter-provincial risk transfer and can explain the risk situation in some non-rice-producing areas.

Health risks under NFS standards and HHRA. Differences among assessment systems may lead to overestimation or underestimation of risks. In this study, the average As concentrations in Chongqing, Sichuan, Shaanxi, Gansu, and Ningxia were 0.082, 0.060, 0.055, 0.099, and 0.072 mg kg^{-1}, respectively. According to the NFS standards, the SFPI values for these five provinces were 0.41, 0.30, 0.275, 0.495, and 0.36, respectively, which were far below the risk threshold (1.0). These results indicate that the As concentration in rice in these provinces is safe. However, when we used HHRA to re-examine the risk imposed by rice As concentration in the five provinces, the average HQs calculated for these provinces were 1.8–3.2 times of the risk threshold. The opposite evaluation results were obtained under the two criteria. Although some elements, such as As and Cr, did not exceed the MAC in rice, their levels could still be high enough to cause non-carcinogenic hazards, which is consistent with the finding of Lu et al.[38]. By contrast, the Pb and Hg concentrations exceeded the MAC, but they caused no non-carcinogenic hazard. The Pb concentrations in Tianjin, Anhui, and Jiangsu were 0.25, 0.25, and 0.29 mg kg^{-1}, respectively. According to the NFS standards, their SFPI values were 1.25, 1.25, and 1.45, respectively. According to HHRA conducted to measure the risk, the HQs were 0.52, 0.56, and 0.64, respectively.

Due to different situations in various countries, the angles considered in the standard formulation process, and the critical protection objectives, there are universal differences in standards. The primary purpose of food safety standards is to ensure human health, which requires strengthening of the simulation and evaluation of localized exposure. Because rice will still be the staple food and a major source of nutrients for a long time, the long-term goal of risk reduction should be the reduction of heavy

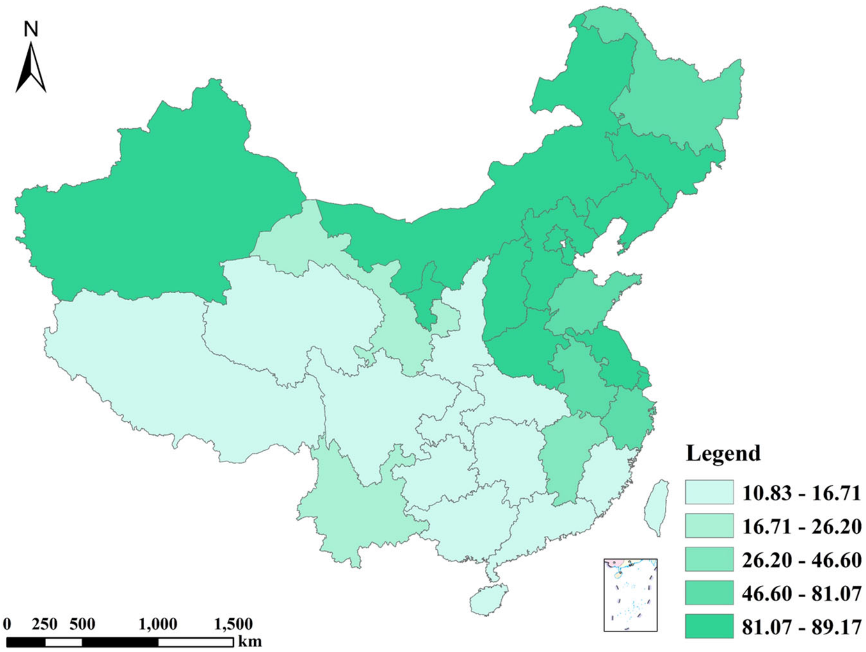

Fig. 5 Rice quality–heavy metal(loid) (RQHM) score distribution. [Note: the shade of green is used to represent the RQHM score. The darker the green, the higher the score].

metal(loid) concentrations in rice. In addition, heavy metal(loid) concentration is not the only parameter affecting the risk. Consideration of the dietary characteristics of the population in the target area and setting of pollutant limits according to local conditions could effectively ensure human health and avoid the waste of resources. In summary, we suggest to update and subdivide the body weight and intake parameters of exposed populations and incorporate parameter differences into limit criteria to ensure food safety and human health.

Limitations and future directions. Similar to other studies, several issues remain be addressed to develop a more sophisticated approach for health risk assessment. Future work can include but not be limited to the following areas for more complete protection of human health. Firstly, only the average intake level of different age groups in each province was considered in this study, which can hardly reflect the individual differences and variations. In the future, the national nutrition and health survey data should be integrated to improve the assessment methods to more accurately evaluate the individual intake differences and health risks. Secondly, this study only evaluated rice and related problems. Since rice is one of many foods and we did not consider other food varieties, this study might have actually underestimated the health risks of heavy metal(loid)s. The risk of dietary intake should be more comprehensively assessed in the future. Thirdly, we only examined Cu and Zn as metals with nutritional value. In fact, many factors affect the nutritional value of rice, such as climate change[44–46], farming practices, and rice varieties[47]. In the future, the health benefits of climate and optimal field management should be considered.

Conclusions

This study revealed that the heavy metal(loid) concentrations in commercial rice generally met NFS standards in China. However, there could still be health risks for certain critical receptors, such as toddlers (2–5 years) and children (5–12 years). Through a probabilistic risk assessment, we found that there are still health risks when the heavy metal(loid) concentrations are lower than the MACs. There are still relatively high non-carcinogenic health risks for critical populations in central and southern China. Arsenic contributes the most to the overall health risk (2.8–100%), followed by Cd (0–96.81%). Both body weight and rice intake have an impact on the final risk. The fuzzy evaluation results indicated significant regional differences in the safety and quality of rice in China due to the presence of heavy metal(loid)s. In south China, measures are needed to reduce the risks from heavy metal(loid) intake due to rice consumption.

Importantly, uniform parameters were replaced with refined exposure parameters for risk assessment in this study. We identified critical receptors in 32 provinces and revealed mismatch between NFS standards and human health risks. The results suggest that policymakers should adopt local measures to reduce the concentrations of heavy metal(loid)s in rice to protect human health.

Methods

Data collection. Data were obtained from the Web of Science and China National Knowledge Infrastructure databases. By using “heavy metals”, “rice”, “risk assessment”, and “China” as the keywords, 1182 peer-reviewed articles on heavy metal(loid) concentrations in rice published from 1997 to 2021 were collected. We then identified and removed duplicate articles mostly based on the title, abstract, and keywords. To achieve reliable heavy metal(loid) risk assessment, the research focused on particular areas, such as mining and sewage irrigation areas, was excluded. In addition, the NFS standards issued by the Chinese government, including the determination of cadmium (Cd) (GB 5009.15-2014) and lead (Pb) (GB5009.12-2017) concentrations in food, were used in this study. Finally, 3376 heavy metal(loid) concentration data points were obtained from 408 articles (Supplementary Fig. 1).

Single factor pollution index assessment. Single factor pollution index (SFPI) can reflect the degree of pollution of various heavy metal(loid)s, and is the pollution assessment method most often used in China. Here, this method was chosen to allow direct comparison with the results of previous studies. The formula can be expressed as

$$P = \frac{C}{\mathrm{MAC}}, \tag{1}$$

where P is the single factor pollution index of heavy metal(loid), C is the concentration of the heavy metal(loid), and MAC is the standard concentration of the heavy metal(loid). A P value < 1.0 indicates that the element is at a safe concentration, while P > 1.0 represents that the concentration of the element exceeds the NFS standard. With an increase in *P value*, the cumulative amount also increases.

Human health risk assessment. The potential health risks of heavy metal(loid)s were assessed using HHRA as recommended by the United States Environmental Protection Agency (2011). This method allows the consideration of differences in region and dietary habit to obtain more accurate results. Heavy metal(loid)s have different non-carcinogenic and carcinogenic effects on human health. In this study, only the health risks generated by the oral consumption of rice were evaluated. The whole population of China was the study target and was divided into four age groups. Supplementary Table 1 summarizes the age composition and exposure parameters of each age group. The average body weight and daily intake were determined according to the "Handbook of Exposure Parameters for the Chinese Population" issued by the Ministry of Environmental Protection of China. The average daily dose (ADD) was used as an exposure metric to estimate adverse health effects, which was quantified by intake dose, body weight, and average time:

$$\mathrm{ADD} = C \times \mathrm{IR} \times \mathrm{ED}/(\mathrm{BW} \times \mathrm{AT}), \tag{2}$$

where C is the heavy metal(loid) concentration in rice (mg kg^{-1}), IR is the daily rice intake (kg day^{-1}), ED is the exposure duration (days), BW is body weight (kg), and AT is the average time (days). According to the model guidelines, human health risks can be categorized as carcinogenic risk (CR) and non-carcinogenic risk (NCR). NCR was expressed by comparing the ADD with the reference dose (RfD). The ratio of the ADD to RfD can be expressed as the hazard quotient (HQ):

$$\mathrm{HQ} = \mathrm{ADD}/\mathrm{RfD} \tag{3}$$

The CR in different age groups was calculated as the ADD of each age group multiplied by the appropriate oral cancer risk slope factor (SF). Because the estimation of the carcinogenic potential of carcinogens is based on the assumption of lifetime exposure, the incremental lifetime cancer risk (ILCR) was calculated by summing the CR of different age groups using the direct arithmetic weighting method:

$$\mathrm{CR} = \mathrm{ADD} \times \mathrm{SF} \tag{4}$$

$$\mathrm{ILCR} = \sum \mathrm{CR} \times F, \tag{5}$$

where *CR* represents the CR of an age group, and *F* represents the proportion of the corresponding age group. The RfD and SF values of the five heavy metal(loid)s (Cd, As, Hg, Pb, and Cr) were obtained from the literature or authoritative chemical toxicity databases. Supplementary Table 2 lists the contaminant limits in foods in China's NFS standards and the reference intakes of dietary nutrients for Chinese residents.

Probabilistic assessment. Monte Carlo simulation was used evaluate the uncertainty of parameters in health risk assessments. Due to data limitations, we used the Monte Carlo simulation method to simulate the data distribution and ensure that the results could reflect the actual situation. The heavy metal(loid) concentration data fitted a log-normal distribution (Supplementary Table 3). Moreover, we fitted the optimal probability distribution of other exposure parameters (Supplementary Table 1). First, the probability of the NCR exceeding the risk threshold (P_{NCR}) was determined. If HQ > 1.0, there is a potential health risk. It is essential to ensure that sensitive receptors are included in the distribution used for receptor characteristics. Therefore, the highest P_{NCR} calculated for different age groups was selected to evaluate the NCR. Furthermore, the probability of the ILCR exceeding the risk threshold (P_{ILCR}) was determined. ILCR values above 1×10^{-4} were considered to represent an unacceptable CR, while values below 1×10^{-6} were considered to represent a negligible CR. For the five heavy metal(loid)s (Cd, As, Hg, Pb, and Cr), high values of P_{NCR} and P_{ILCR} were considered to represent the probability of exceeding the risk threshold (P_{HR}) using the maximum operator:

$$P_{\mathrm{HR}} = \mathrm{Max}\,(P_{\mathrm{NCR}}, P_{\mathrm{ILCR}}) \tag{6}$$

$$P_{\mathrm{NV}} = \mathrm{Max}\,(P_{\mathrm{Cu}}, P_{\mathrm{Zn}}) \tag{7}$$

The effects of Cu and zinc (Zn) on the nutritional value of rice were also evaluated using the probability method, and the probability of exceeding the corresponding nutritional value impact (P_{NV}) was determined. The P_{NCR}, P_{ILCR}, and P_{NV} values were processed using fuzzy techniques.

Fuzzy assessment. To explain the excess probability generated by the probability assessment, a fuzzy membership function was developed to systematically convert human perception and language variables into values. Five grades were established to express the critical level of standard exceedance probability: "high" (H), "medium–high" (MH), "medium" (M), "low–medium" (LM), and "low" (L). As shown in Supplementary Fig. 2, fuzzy membership functions were used to represent language variables. The P_{HR} and P_{NV} obtained by the probability analysis were mapped to the membership function to generate the fuzzy critical level. The fuzzy rice quality heavy metal(loid) (RQHM) score was obtained using fuzzy logic operators and rule aggregation. Fuzzy logic operators included AND and OR:

$$\mathrm{AND}: \mu^{\mathrm{RQHM}} = \mathrm{Min}\,(\mu^{\mathrm{HR}}, \mu^{\mathrm{NV}}) \tag{8}$$

$$\mathrm{OR}: \mu^{\mathrm{RQHM}} = \mathrm{Max}\,(\mu^{\mathrm{HR}}, \mu^{\mathrm{NV}}) \tag{9}$$

As shown in Supplementary Table 4, 25 fuzzy rules were developed to qualitatively determine RQHM. Fuzzy health risks and nutritional effects can be aggregated using the AND operator. These combinations led to four different fuzzy RQHM results. These results could be mapped into a new set of fuzzy membership functions, as shown in Supplementary Fig. 3. Five grades were established to describe RQHM: "excellent" (E), "good" (G), "general" (M), "poor" (P)", and "very poor" (VP). The OR operator was used to aggregate the resulting fuzzy RQHM and generate the final fuzzy RQHM membership function. It was defuzzified to obtain an RQHM score in the range of [0, 100]. The higher the score, the better the RQHM. The gravity-based centroid of the final fuzzy RQHM membership function was determined as the final RQHM score:

$$\mathrm{Centroid} = \int x\mu_A^x \mathrm{d}x / \int \mu_A^x \mathrm{d}x \tag{10}$$

The framework of integrating probabilistic and fuzzy methods to evaluate rice food safety is shown in Supplementary Fig. 4. The results of the probability assessment included the P_{HR} and P_{NV}. At the same time, a fuzzy assessment could provide a score for rice food safety assessment. The RQHM score can be used to compare the quality of rice under the influence of heavy metal(loid)s, as well as to support the decision to adopt appropriate heavy metal(loid) control measures to improve the safety level of rice.

Data availability

The datasets generated during the current study are available at https://figshare.com/s/f77438019573046c05a6.

Received: 18 October 2022; Accepted: 21 February 2023;
Published online: 23 March 2023

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Acknowledgements
This study was supported by the National Natural Science Foundation of China (No. 42107509), National Natural Science Foundation of Hubei Province, China (2020CFA013) and Postdoctoral Innovation Research Position of Hubei Province.

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R.W.: investigation, conceptualization, data curation, methodology, visualization, and writing- original draft. C.C.: conceptualization, methodology, resources, visualization, and writing – review and editing. M.K.: data curation. Z.L.: conceptualization and methodology. Z.W.: conceptualization, resources, and writing – review and editing. J.C.: resources. W.T.: supervision, funding acquisition, project administration, and writing – review and editing.

Competing interests
The authors declare no competing interests.

Additional information
Supplementary information The online version contains supplementary material available at https://doi.org/10.1038/s43247-023-00723-7.

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Peer review information *Communications Earth & Environment* thanks Md. Abu Bakar Siddique, Eliaza Jones Mkuna and the other, anonymous, reviewer(s) for their contribution to the peer review of this work. Primary Handling Editors: Sadia Ilyas and Clare Davis. Peer reviewer reports are available.